

DISCUSSION OF THE AMENDMENT

The specification has been amended to provide antecedent basis for Claims 3 and 4.

Claim 1 has been amended, as supported in the specification at the sentence bridging pages 11 and 12. Claim 9 has been amended by deleting the twice-recited term “preferably”

No new matter is believed to have been added by the above amendment. Claims 1-10 remain pending in the application.

REMARKS

The rejection of Claims 1-10 under 35 U.S.C. § 103(a) as unpatentable over US 3,829,505 (Herold) alone or in view of EP 0759450 (Lawrey et al), is respectfully traversed.

As recited in above-amended Claim 1, an embodiment of the present invention is a process for preparing polyether alcohols comprising reacting saturated alkylene oxides with at least one saturated OH compound, or an alkylene oxide that has previously been oligomerized or polymerized with the saturated OH compound, in the presence of a double metal cyanide (DMC) catalyst, wherein an antioxidant is added before the reaction, using more antioxidant than DMC catalyst (based on the mass of DMC catalyst used), wherein the amount of antioxidant is from 2 to 500 times the mass of catalyst used.

Thus, in the presently-claimed invention, the antioxidant is added **before** the reaction, and it is used in an amount of **at least 2 times the mass of the catalyst used**. The significance of these limitations is demonstrated in the specification herein in Examples 1-11, beginning at page 12, line 26. The results of these examples are shown in Table 1 at page 15 of the specification, reproduced below.

Table 1

Example	Amount of DMC [ppm]	Stabilizer	Amount [ppm]	Induction time (min)
1	150	-	-	14
2	150	BHT	1000	4
3	150	BHT	1000	5
4	150	alpha-Tocopherol	1000	5
5	150	Dihexadecylhydroxylamine	500	7
6	150	Triphenyl phosphite	500	4
7	150	Diphenyloxamide	2000	2
8	25	-	-	27
9	25	BHT	250	6
10	25	BHT	250	5
11	25	BHT	50	10

The examples clearly show that adding a sufficiently high amount of antioxidant significantly shortens the induction time. The greater the amount of antioxidant added before the reaction with respect to the amount of catalyst used, the shorter the induction time. Example 8, which is a comparative example, contains no antioxidant (stabilizer), and has an induction time of 27 minutes. Example 11 employs an antioxidant in an amount of twice the amount of catalyst, and has an induction time of 10 minutes. Examples 9 and 10 employ the antioxidant in an amount of 5 times the amount of catalyst, and have an induction time of 5 or 6 minutes.

Herold discloses in his process that antioxidants or antidegradants, such as phenyl beta naphthylamine (PBNA), are desirably added prior to or after polymerization to avoid degradation that might occur, and that PBNA may be used in an amount approximately equal to the amount of the catalyst during telomerization, although some antidegradants may retard polymerization and should be added after telomerization (column 6, lines 46-53). Thus, Herold discloses using an amount of antioxidant approximately equal to the amount of catalyst, i.e., an amount of antioxidant lying between Examples 8 (no antioxidant) and 11 (two times the amount of DMC catalyst) of the invention. In effect, Applicants have compared to the closest prior art, i.e., Herold, contrary to the finding by the Examiner.

The Examiner finds that Applicants should have compared to example(s) obtained by adding antioxidants in amounts disclosed by Lawrey et al prior to polymerization as disclosed by Herold. However, this clearly is not the law. See MPEP 716.02(e) and the cases cited therein, especially *In re Chapman*, 357 F.2d 418, 148 USPQ 711 (CCPA 1966) (**copy enclosed**) (Requiring applicant to compare claimed invention with polymer suggested by the combination of references relied upon in the rejection of the claimed invention under 35 U.S.C. 103 “would be requiring comparison of the results of the invention with the results of the invention.” 148 USPQ at 714.)

While Lawrey et al discloses an amount range of antioxidant (page 4, lines 47-49), it is based on the polyol (polyether alcohol) weight, and is not tied to the amount of DMC catalyst. In addition, Lawrey et al is silent as to when exactly the antioxidant is added to the polyether alcohol. Example 5 of Lawrey et al is the only example that illustrates polyether alcohol preparation using the DMC catalyst, but it contains no disclosure about adding an antioxidant, let alone when it is added. Figure 4 thereof is disclosed is showing the effect of various residual catalyst concentrations of BHT stabilized polyether alcohol (page 7, lines 4-5), but there is no disclosure of when the BHT is added.

Thus, it is clear that Herold is the closest prior art. In addition, and as discussed above, in disclosing that some antidegradants may retard polymerization and should be added after telomerization, Herold actually teaches away from the present invention.

For all the above reasons, it is respectfully requested that this rejection be withdrawn.

The objection to the specification is now moot in view of the above-discussed amendment. Accordingly, it is respectfully requested that the objection be withdrawn.

All of the presently-pending claims in this application are now believed to be in immediate condition for allowance. Accordingly, the Examiner is respectfully requested to pass this application to issue.

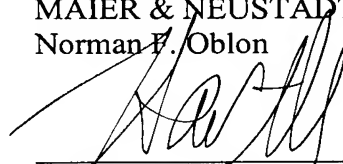
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In re Chapman and Cosby, 148 USPQ 711 (CCPA 1966)

In re Chapman and Cosby

**(CCPA)
148 USPQ 711**

**Decided Mar. 17, 1966
Appl. No. 7412
U.S. Court of Customs and Patent Appeals**

Headnotes

PATENTS

1. Patentability-Composition of matter (§ 51.30)

Patentability - Invention - Specific cases-Chemical (§ 51.5093)

It cannot be said that all products (compositions of matter) of obvious processes are unpatentable by reason of their being inherent results of those processes.

2. Patentability - Anticipation - Patents-In general (§ 51.2211)

Reference patent can be used for all it realistically teaches and is not limited to disclosures in its specific illustrative examples.

Particular patents-Polyethylenes

Chapman and Cosby, High Strength Chlorinated Polyethylenes and Process of Preparing Them, claims 1 to 6 and 8 of application refused.

Case History and Disposition:

Appeal from Board of Appeals of the Patent Office.

Application for patent of Wilbur F. Chapman and John N. Cosby, Serial No. 819,106, filed June 9, 1959; Patent Office Group 140. From decision rejecting claims 1 to 6 and 8, applicants appeal. Affirmed.

Attorneys:

Elizabeth Hunter and George B. Campbell, both of New York, N.Y., and Arnold B. Christen, Washington, D.C., for appellants.

Clarence W. Moore (Raymond E. Martin of counsel) for Commissioner of Patents.

Judge:

Before Worley, Chief Judge, and Rich, Martin, Smith, and Almond, Associate Judges.

Opinion Text

Opinion By:

Rich, Judge.

This appeal is from the decision of the Patent Office Board of Appeals affirming the examiner's rejection of product claims 1 to 4 and process claims 5, 6 and 8 in application serial No. 819,106, filed June 9, 1959, for "High Strength Chlorinated Polyethylenes and Process for Preparing Them." No claim is allowed.

Appellants invented improvements in the art of chlorinating polyethylenes. Their application points out that while it is known to chlorinate polyethylenes having a wide variety of structures and molecular weights to obtain resins useful in many applications, such prior art chlorinated polyethylenes are "generally lacking in the high strength characteristics required for the purposes served by the more expensive, rubber-like, thermoplastic resins of the polyvinyl and polyvinylidene chloride types." This lack has been due, according to appellants, in part to an inherent lack of these properties in the polyethylene materials chlorinated and in part to degradation of the polyethylene molecules during chlorination, resulting in products of low molecular weights.

According to appellants' brief, by the "combined factors of using as starting materials high molecular weight polyethylenes which themselves have high tensile strengths, and subjecting these polyethylenes to chlorination under such conditions as to minimize molecular weight

degradation and consequent loss of strength characteristics" appellants have produced chlorinated polyethylenes alleged to possess "outstandingly high [tensile] strength characteristics * * *, rendering the resulting products useful in the production of films, coatings, floor coverings and wall coverings of high strength and durability." Appellants emphasize the importance of maintaining the rate of chlorination "within relatively narrow limits" to minimize molecular weight degradation.

Claims 1 and 5, to the product and process, read:

1. A chlorinated polyethylene resin having a chlorine content between about 25% and about 65% by weight, molecular weight characteristics such that its intrinsic viscosity as measured in o-dichlorobenzene at 100° C. is not less than about 1.8, chemically inert, insoluble in organic solvents at 20-25° C., having a tensile strength value according to ASTM method D-638-58T of at least about 3,000 and equal to at least about 100 times the weight percent chlorine in the resin, having a true ultimate tensile value of at least about 8,000, and having an infrared spectrogram showing characteristic absorption peaks at the following wave length: 3.42 to 3.5 microns; 3.38 to 3.48 microns; 6.8 to 6.9 microns; and at 7.8 to 7.9 microns.

5. A process for producing chlorinated polyethylene resins of high strength characteristics which comprises contacting, in finely divided porous powder form, a polyethylene having a weight average molecular weight between about 1,000,000 and about 5,000,000, density between about

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0.935 and about 0.985, with a chlorine-containing gas containing chlorine and an inert gas in a ratio of not less than about 0.5 part of inert gas per part of chlorine by weight, at temperatures between about 40° C. and about 100° C., and continuing the contacting until the resulting chlorinated product contains between about 25% and about 65% chlorine by weight, said conditions of temperature, chlorine to inert gas ratio and degree of chlorination being so correlated that the rate of chlorination in terms of weight gain per hundred unit weights of reacting charge per hour is between about 0.5 and about 15.0.

Claims 2, 3 and 4 are dependent on claim 1 and recite further limitations such as an absorption peak at 13.88 microns (claims 2 and 3), a narrower range of chlorine content in the resinous products (claim 2), an intrinsic viscosity range between about 4.0 and about 3.0 (claim 2), higher lower limits for tensile strength and ultimate tensile strength (claim 2), intensity values for certain absorption peaks (claim 3), and the existence of the resin in "particulate form" (claim 4), the particles having densities within a range of about 1.15 and about 1.65.

Claim 6 is dependent on claim 5 and calls for maintaining the temperature between about 40° C. and about 50° C. until the chlorine content is between about 20% and about 30% and then raising the temperature and maintaining it between about 60° C. and about 90° C. until a chlorine

content of about 45% to about 65% has been reached.

Claim 8 is independent and defines a method for producing a chlorinated polyethylene having the chlorine content and strength characteristics of the resin defined by claim 2, by a process such as that defined by claim 5.

The references relied on are:

Hoerger et al. 2,913,449 Nov. 17, 1959

Noeske 2,928,819 Mar. 15, 1960

Noeske is entitled "Process for the Chlorination of Polyethylene" and discloses a process for treating solid, powdered, low molecular weight (i.e., about 100,000 to about 300,000) polyethylene, at temperatures up to about 110° C., with gaseous chlorine diluted, if desired, with inert gases. Noeske admixes a pulverulent inert-to-chlorine material such as magnesia, anhydrous magnesium sulfate, anhydrous aluminum sulfate, anhydrous sodium sulfate, or sodium chloride, with the powdered polyethylene prior to chlorination, thereby permitting the use of higher reaction temperatures and consequent shorter reaction times, without entailing the risk of charring the product because of the evolution of excessive heat. Noeske says that prior to his work it had been difficult to obtain products having a chlorine content above about 40% by weight because extremely long chlorination periods were required. The degree of chlorination disclosed by Noeske ranges from 52% in example 1 to 69% in example 6. The actual temperatures employed range from 50° C. in example 7 to 105° C. in example 6. In six of Noeske's eight examples, a constant weight ratio of nitrogen to chlorine of 0.4 is employed; and, in the other two, a nitrogen to chlorine ratio by weight ranging initially from 2.0 to 0.5 after five hours is used. The highest per cent chlorination at the end of this period appears to be about 32, the reaction temperature being about 65° C. throughout.

Hoerger discloses a process for chlorinating a high molecular weight polyolefin by a so-called "conjoint halogenation technique." Finely divided powdered polyethylene sequentially undergoes "direct" chlorination with a mixed stream of chlorine and nitrogen, this step ordinarily being employed first, and a "solution chlorinating" step. The chlorination of polyethylene, polypropylene, polybutylene, and other polyolefins having linear, unbranched molecular structures is disclosed, such starting materials having molecular weights "usually from at least 40,000 to as much as 500,000 and 3,000,000 and greater." Specifically disclosed is the *direct* chlorination of *polyethylene*, "especially that having a relatively high molecular weight of from 40,000 to 500,000 and greater * * * by subjecting the agitated, freeflowing mass of polymer to an atmosphere of chlorine at a temperature between about room temperature and about 85° C." to provide a product having "any desired halogen content to as great as about 80 percent or more by weight * * *."

It is unnecessary to discuss Hoerger's "solution chlorination" process beyond noting that chlorinated polyethylenes obtained by such a process have "a more uniform distribution of the substituent chlorine atoms along the polymer molecule" than is usually the case, and also have

"greater elongations and elasticities than may be obtained in products chlorinated by other methods."

The Process Claims

The examiner's position is thus stated in his answer:

Noeske shows all of the features

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of the process claims, except for the molecular weight of the starting polyethylene. With respect to said starting materials, Noeske contemplates treating polyethylenes prepared by the Ziegler process (column 1, lines 41-50) which process is known to produce polyethylenes of the claimed density. The molecular weights are not held to be critical by Noeske and the highest molecular weight shown is 300,000 (Example 8) apparently determined viscosimetrically as in Example 1. In Example 8, (compared with claim 5 on appeal) the powdered (see claim 1) polyethylene is treated at 80°C for 12 hours with a gas containing chlorine and initially 5 parts by volume of nitrogen per volume of chlorine or about 2 parts by weight. Based on the table at column 4 lines 15-19, the rate of chlorine addition is on the order of 4% per hour during the first 12 hours. At the end of 12 hours the chlorine content is 43% and at 20 hours 61%, both contents falling within the scope of applicants' claim 5. Claim 6 which requires the initial chlorination to be performed at between 40 and 50° C and later between 60 and 90° C is deemed anticipated by the raising of the chlorination temperature in later stages of chlorination in Examples 1-3, 5, 6 and 8 of the reference. The determination of the exact temperatures to use in each stage is deemed well within the skill of the art. Claim 8 is further limited to a chlorine content of 45%-55%, but this is anticipated by the chlorine content of 53% after 16 hours in Example 8 and by the final products of Examples 1, 2 and 3.

It is admitted that Noeske does not specifically disclose the treatment of polyethylenes having weight average molecular weights of as high as 1,000,000. However it is deemed obvious to treat other known polyethylenes (the instant polyethylenes have not been alleged to be novel) by the process of Noeske. This conclusion is deemed further supported by Hoerger et al. who disclose the chlorination (in another manner) of polyolefins including polyethylenes having a molecular weight as high as 3,000,000 (column 1 lines 50-61). It is deemed obvious to substitute the olefins of Hoerger et al. for those of Noeske.

Without passing on the examiner's finding that certain claims are "anticipated," we otherwise agree with his reasoning and ultimate conclusions, adopted in essence by the board. We also express our agreement with the examiner and board, by way of responding to appellants' contention that Noeske requires the presence of an added solid pulverulent diluent substance, that (1) appellants' claims are sufficiently broad to include the use of such a substance and (2) portions of Noeske's examples 1 and 7 disclose processes (for comparison purposes) wherein the

added pulverulent material is omitted.

We have considered appellants' remaining arguments, many of which go to the question of novelty, not obviousness, but are unable to agree that the board's decision should be reversed. Their most noteworthy contention, we think, is:

Although the differences in Noeske's and applicants' conditions may appear slight, they spell the difference between failure (Noeske), who obtained a charred product in the absence of powdered diluent, and success by the applicants procedure * * *. The Noeske disclosure thus leads away from the applicants' process of chlorination, which does not depend upon the presence of added inert material, yet may be performed at temperatures as high as 100°C. with his controlled chlorine dilution.

We think this argument overlooks the fact that one rejection is based on both Noeske and Hoeger. While it might be surprising, in view of the Noeske disclosure, to find *no* charring if *low* molecular weight polyethylene were directly chlorinated at temperatures of about 60°C. and above without the use of a pulverulent diluent, this is not appellants' invention. Whether one skilled in the art, looking at Noeske *alone*, might expect the product to char if a polyethylene of 1,000,000 to 5,000,000 molecular weight were used, we need not say, for we believe the total disclosure of Noeske and Hoeger suggests that it would not.

The Product Claims

Notwithstanding the solicitor's urging that product claims 1 to 4 should stand or fall with process claims 5, 6 and 8, and that all claims should stand or fall with process claim 5, we consider the question of the patentability of those products to be a separate issue, to which we now turn.

We again consider the rejection on the disclosures of both Noeske and Hoerger. The examiner's answer says:

It is deemed that in view of the similarity in preparation, the claimed polymers must be the same as the Noeske polymers except for prop

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erties that are the result of treating a higher molecular weight starting material. Ex parte Fryling et al., 723 OG. 5, 115 USPQ 99. Applicants, in an affidavit submitted February 9, 1962 have shown certain properties * * * for a polymer ¹ having a molecular weight of 170,000 chlorinated in the manner taught by Noeske. Of the properties compared in the affidavit, ultimate elongation does not appear in the claims, ultimate tensile strength falls somewhat short of the claimed limitation, "equal to at least about 100 times the weight percent chlorine content" and the true [ultimate] tensile strengths of 8,677 and 12,080 compared with "at least about 8,000" in claim 1 and "at least about 10,000" in claim 2. The claims also recite an intrinsic viscosity limitation, and since this corresponds to

molecular weight, it is presumably not met by the lower molecular weight polymers of Noeske.

In view of the fact that the polyethylene of the affidavit is lower in molecular weight than the highest molecular weight polyethylene of Noeske and in view of the use of "about" with respect to the limitations of the claims the difference between the properties of the Noeske polymer and the properties specified in the claims is not apparent. At any rate, the differences are not considered to be differences in kind and have not been shown to be due to a factor other than the use of a higher molecular weight starting material, which is obvious as explained with respect to the rejection of the process claims.

The board added:

* * * the affidavit is silent as to the results obtained when the high molecular weight polyethylene of Hoerger et al. is substituted for the polyethylene of Noeske. This is the essence of the examiner's rejection and it would appear that such a substitution would inherently yield a product substantially the same as that claimed.

[1] We do not agree with the board that a high molecular weight polyethylene of Hoerger should have been substituted for the polyethylene of Noeske if comparative data are to be presented for this, we think, would amount to requiring comparison of the results of the invention with the results of the invention. Nor can we agree that appellants' compositions are unpatentable because such a process "would inherently yield" a product substantially the same as that claimed, since that position implies that any and all products of obvious processes are unpatentable by reason of their being the "inherent" results of those processes. The issue is simply obviousness of a composition of matter and we do agree with the board's decision on that issue.

In describing his *directly* chlorinated products, Hoerger says the resultant chlorine distribution in the polymer chain is "more intermittent, non-statistical * * * block-like," and states that "as many as four adjacent methylene groups may be found at random points in the polymer molecule." Products obtained by the "direct" method are said to "have *greater tensile strengths and rigidities* " than products obtained by other methods. (Emphasis ours.) Hoerger goes on to say:

The halogenated and particularly the chlorinated products of the present invention have felicitous typical properties and characteristics which usually include good elastomeric propensities, generally satisfactory elongations and excellent tensile strengths. Although the properties of the various products that may be obtained by practice of the duplex halogenating procedure of the present invention are dependent and tend somewhat to be patterned after the properties attributable to the predominant type of substitution that is effected, they are not necessarily average or linearly proportionate values that are strictly predictable on the percentage of combined chlorine or other halogen that has been attached during either the direct or the solution step of the total halogenation procedure.

This point is further illustrated in the subsequent examples contained herein. In this connection, however, particularly desirable halogenated products, especially chlorinated products, may usually be prepared when the predominant proportion of the total combined halogen has been attached by block distribution - favoring direct halogenating techniques. *Such products ordinarily are characterizable in having high tensile strengths and desirable, moderate degrees of elongatability .* (Emphasis ours.)

By using the conjoint halogenation technique, Hoerger obtained products having tensile and elongation properties intermediate those of the product obtained through use of "direct" or "solution" chlorination processes alone.

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In response to the examiner's rejection, appellants submitted a Rule 132 affidavit which they stated supported their position that the claimed process "is patentably different from those of either of the references taken alone or together and produces a different product." The examiner's answer, quoted above, gives his analysis of that affidavit. He observed, inter alia, that "the polyethylene of the affidavit is lower in molecular weight than the highest molecular weight polyethylene of Noeske * * *." The examiner was there referring to the fact that appellants chlorinated a polyethelene of 170,000 molecular weight in accordance with the procedure of Noeske's Example 8 whereas that example employs a polyethylene of 300,000 molecular weight. The examiner also noted that the claimed polymers and the polymers of the affidavit differed in ultimate tensile strength but did not differ in *true* ultimate tensile strength. ²

Considering that appellants, in their specification regard true ultimate tensile strength as a more "true and accurate evaluation" of the absolute strength of the material evaluated than ultimate tensile strength, we think the examiner was correct in concluding that the affidavit was not adequate to establish significant differences in properties between the claimed polymers and those of Noeske. Also, comparing the claimed polymers with the closest polymer of Noeske, i.e. one of 300,000 molecular weight, would have been the most meaningful comparison in all respects and is what, we think, appellants should have done if comparative data were to be relied upon for patentability.

Although appellants say the purpose of their affidavit was to show that the claimed products are different from those of both references, taken alone or together, no comparison was made with the polymers of Hoerger. They do compare Hoerger's polymers with theirs in their brief, saying:

The highest tensile product disclosed by Hoerger et al. is 2,580 psi. in Example 1. This is for a 42 percent chlorine product. Applicants' claim 1 provides for a tensile strength of at least about 3,000 and equal to at least about 100 times that weight percent chlorine in the resin. Thus the applicants' 42% chlorinated resins have tensiles of at least about 4,200. Similarly, Hoerger's Example II, wherein a 43% chlorine resin had a tensile

of only 1,500, whereas applicants' 43% chlorine products have tensiles of at least 4,300. Example III of the Hoerger et al. patent discloses a resin of 35% chlorine with a tensile of 2,900 psi., whereas the applicants' 35% chlorine resins have tensiles of at least 3,500. Example IV of Hoerger et al. illustrates a 25% chlorine resin with a tensile of 1,400 psi., whereas applicants' 25% chlorine resins have tensiles of at least 3,000 psi. These variations of tensile strength with chlorine content are graphically illustrated in curve A of Figure 9 of the instant application * * *. Hoerger et al. do not disclose a chlorinated product containing more than 43% chlorine.

Claim 2 of the present application is directed to the applicants' preferred products containing between about 45% and 55% chlorine and having the highest overall strength properties; namely tensile strengths of at least 5,000 psi. combined with good elongation properties so as to provide "true ultimate tensile" values of at least about 10,000, even at these relatively high chlorine contents. Hoerger et al. clearly fail to anticipate this preferred product. They fail to show a product of the same chlorine content, or of comparable tensile strengths.

[2] What appellants fail to point out is that Hoerger used a polyethylene of 60,000 molecular weight as a starting material in Example 1, much lower than the 500,000 to 3,000,000 molecular weight polyethylene Hoerger elsewhere discloses he can use. Moreover, the 42 percent chlorine product Hoerger prepared in Example 1 is not a product obtained by "direct" chlorination alone. Only two-thirds of that chlorine percentage was introduced by the "direct" process; one-third was introduced by the "solution" process. A similar analysis may be applied to the remaining examples of Hoerger. In view of Hoerger's disclosure that a polymer produced by conjoint chlorination has tensile properties intermediate those produced by "direct" or "solution" processes used alone, we think it would be expected that a 42 percent chlorine product produced by the "direct" process alone would have a much higher tensile strength. As noted by Hoerger,

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the "direct" process can introduce 10-80% chlorine into the polyethylene. A reference can be used for all it realistically teaches, and is not limited to the disclosures in its specific illustrative examples. In re Widmer, 53 CCPA —, 353 F.2d 752, 147 USPQ 518, at 523.

Summarizing, while appellants have urged patentability of their claimed polymers on the basis of selected portions of Noeske and Hoerger, they have overlooked other and more relevant disclosures in those references. Considering all of the evidence of record including the close structural similarity, the similar method of making the polymers, the similar properties and uses, we think the examiner has established a prima facie case of obviousness of the claimed invention. Appellants' evidence and arguments have failed to rebut that case. In re Lohr, 50 CCPA 1274, 317 F.2d 388, 137 USPQ 548.

The decision of the board is *affirmed*.

Footnotes

Footnote 1. Two runs were made, giving polymers containing 47.5% and 56.9% chlorine respectively.

Footnote 2. Appellants state that the true ultimate tensile strength of the material gives a better indication of its strength, since calculation of that tensile strength value takes into account the elongation and resultant reduction in cross-section of the test specimen at break. In other words, true ultimate strength tests take into account the minimum cross-sectional area of test sample *at the moment of rupture* whereas ordinary ultimate tensile strength tests do not.

- End of Case -